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GENERAL DYNAMICS | CONVAIR

Report No. 8926-091

Material - Adhesives - High Temperature Ceramic

Literature Survey and Tensile Strengths

D. S. Pratt, J. E. Shoffner, E. E. Keller

2 March 1959

Published and Distributed
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Abstract

A literature survey pertinent to high temperature ceramic adhesives was conducted. Seven literature references are given in the body of the report. Appendix I of the report gives 101 references taken from the technical literature and Appendix II cites 9 references taken from WADC TR 58-184, August 1958. Tensile tests were made with Univdrsity of Illinois U-1067 composition with indifferent results.

Reference: Pratt, D. S., Shoffner, J. E., Keller, E. E., "High Temperature Ceramic Adhesives," General Dynamics/Convair Report MP 58-475, San Diego, California, 2 March 1959. (Reference attached).

STRUCTURES-MATERIALS LABORATORIES

CONVAIR

A DIVISION OF GENERAL DYNAMICS CORPORATION

SAN DIEGO

REPORT MP-58-475

DATE 2 March 1959

MODEL REA-7038

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TITLE

REPORT NO. MP-58-475

HIGH TEMPERATURE CERAMIC ADHESIVES

PREPARED BY D. S. Pratt
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INTRODUCTION:

New high speed aircraft and missiles now on the drawing boards will have high surface temperatures when in service. The surfaces must be smooth in order to keep this temperature as low as possible. Adhesives offer a method of achieving this by keeping irregularities to a minimum. Organic adhesives have a temperature limitation of approximately 600°F, but since skin temperatures are expected to rise above 600°F, they cannot be considered for long time operation.

Ceramic materials, well known for their ability to withstand high temperature, offer a possible source of high temperature adhesives. Various types of ceramic adhesives, i.e., sodium silicate, sodium aluminate, magnesium oxychloride, aluminum phosphate, and potassium silicate, have been in use for years. These materials fail under conditions of heat and/or humidity and are not suitable for conditions found in high speed aircraft. (7) Therefore, other ceramic compositions must be investigated for a possible high temperature adhesive.

LITERATURE SURVEY:

In the course of a three year investigation by Spriggs (2) (7), over 100 different formulations were tried as a high temperature adhesive for types 302 and 17-7 PH stainless steels. These formulations included ceramic-oxide glassy bonded coatings, cermets with sintered metal bonds, air setting temperature resistant silicates, aluminates, oxychlorides, oxysulphides, and ceramic oxide resin bonded materials. Those specimens requiring heat for curing were fired for twenty minutes under a 50 psi loading at temperatures from 1000°F to 2000°F. Tensile testing was done at room temperature, 600°F, 800°F, and 1000°F on a Tinius Olsen testing machine. Spriggs states that some glassy bonded adhesives, when modified with powdered metal additions develop over 1000 psi tensile strength from room temperature to 1000°F. These values were obtained using a stainless steel screen carrier in the joint area.

The Boeing Airplane Company has developed a metal to metal ceramic adhesive. The Narmco Company is licensed for sale of this item, but at last report, December 10, 1958, was not ready to release samples for study. Mr. Roger Long of the Narmco Research Group, San Diego, stated that he believed the upper limit of the adhesive would be about 800°F; however, the values at that temperature would be over 1000 psi. (3)

Besides the Boeing adhesive, the Narmco laboratories are working on a WADC contract AF 33(616)-5776 and Navy BuAero contract NOas58-587C. The WADC work is a follow-up on the University of Illinois work, while the Navy contract is on the development of an exothermic adhesive.

The Aeronca Company, Middletown, Ohio, is also working on further development of the University of Illinois research. This work is being done under WADC contract AF 33(616)-5538.

DISCUSSION OF LITERATURE SURVEY:

Throughout the University of Illinois investigation it is stated that the best adherence will be achieved when the coefficients of expansion of the metal and the ceramic adhesive are closely matched. However, one of the final compositions developed, UI 1067, has a very low coefficient, 86.1×10^{-7} cm/cm/°C. This adhesive

DISCUSSION OF LITERATURE SURVEY: (Continued)

is as favorably reported as another which has an expansion factor of 423×10^{-7} cm/cm/°C. Stainless steel alloy, type 302 has a coefficient of 518×10^{-7} cm/cm/°C. This laboratory is unable to explain why this formulation was considered for investigation or why it functions as it does.

The function of the stainless steel carrier is unexplained. It was proven to be an aid in achieving bond, but just how it does was not discussed. A fiberglass carrier in organic adhesives localizes shrinkage, maintains a predetermined joint spacing, and prevents the propagation of cracks. Does the stainless steel carrier function in the same manner, or is there a union of metals at the carrier-metal interface that gives added strength?

Some work was done with the addition of adherence oxides to the adhesives, but this line of investigation was limited. No formulations were tried with cobalt oxide which is known to contribute greatly to the bonding of porcelain enamel to metal. (4)

Nearly all of the University of Illinois work was done on metal roughened by sand blasting or other mechanical means. Another method of metal preparation, nickel plating, was investigated to a smaller extent.

INITIAL LABORATORY STUDY OF UNIVERSITY OF ILLINOIS MATERIAL:

Adhesive Frit Preparation - The University of Illinois composition UI 1067 was favorably reported by Spriggs. (7) This formula was used to compound a frit for the initial laboratory study.

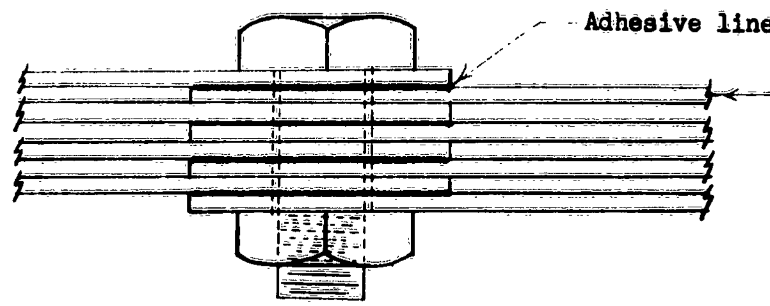
Adhesive Frit Oxide Composition Parts by Weight		Frit Batch Composition Parts by Weight	
SiO_2	38.0	Quartz	24.8
Na_2O	5.0	Sodium Nitrate	9.0
B_2O_3	57.0	Boric Acid	66.2

The frit was ball milled with silica and water until the residue on a 200 mesh screen was less than 2% of the weight of a 50 cc sample.

Adhesive Frit Ball Mill Formula Parts by Weight	
Frit UI 1067	100
Colloidal Silica	4
Water	28.2

INITIAL LABORATORY STUDY OF UNIVERSITY OF ILLINOIS MATERIAL: (Continued)

Adhesive Slip Application and Curing - Sandblasted specimens of type 321 stainless steel were sprayed with the adhesive slip until a dried thickness of 7-10 mils was achieved. They were dried at 200°F for one hour. The specimens were assembled to give a net contact area of 1.3 in^2. They were pressed together as tightly as possible by means of a 1/2" nut and bolt arrangement, and fired at 1750°F for twenty minutes without the application of additional pressure.



Method of fastening specimens

Tensile Shear Test Results - Of the eight joints compressed and cured, only three survived the disassembling of the nut and bolt. The oxide scale formed between the nut and bolt made the disassembly very difficult. The specimens were pulled on a Tinius Olsen tensile machine at room temperature. The loading rate was 600 ± 10 lbs. per minute.

Joint No.	Tensile Value
1	Broke on Disassembly
2	" " "
3	215 psi
4	529 psi
5	Broke on Disassembly
6	330 psi
7	Broke on Disassembly
8	" " "

Average 358 psi

DISCUSSION OF INITIAL LABORATORY STUDY:

The average tensile value of 358 psi is lower than the 545 psi obtained by the University of Illinois; however, the conditions were not the same. The University of Illinois applied a 50 psi loading during the curing and cooling, and used a 28 mesh stainless steel screen as an adhesive carrier. Our initial evaluation seems to indicate that the values reported by Spriggs (7) can be obtained if all conditions are duplicated. Duplication will require the development of a fixture to apply the necessary loading during firing and utilizing 28 gauge stainless steel screening.

RECOMMENDATIONS FOR FUTURE PROGRAM:

A. The first efforts should be to check the reproducibility of the University of Illinois investigation. In addition to checking with stainless steel alloys 302 and 17-7 PH, superalloys of the Rene 41 and Hastelloys R235 or 25(L605) types would be used. The latter alloys are ones being considered for the fabrication of new high performance aircraft and missiles, and the adhesives must be applicable to them. Other frit compositions should be studied.

B. The types of bond achieved would be studied by metallographic, x-ray, and electron microscopic methods. After determining the mechanism of bonding, a study should be made of possible changes in the formulas to improve the adhesive character of the bond.

C. An investigation should be made into various methods of metal preparation. The introduction of another strata into the system, i.e., metal plating, may bridge the difference in expansion between the metal and the adhesive to a degree where it would be less critical. This strata also might provide a better bonding surface for the ceramic adhesive.

D. After developing a ceramic adhesive that would give the desired results in one type of testing, i.e., tensile shearing, an investigation should be made of other possible design parameters. Values would be obtained from twist, peel, compression and impact types of testing. Information of this type would be of value to design and other groups that would have the need to know the intrinsic properties of ceramic adhesives.

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APPENDIX I

A LITERATURE SURVEY ON CERAMIC TO METAL ADHERENCE

The effectiveness of a high temperature ceramic adhesive will depend on the union achieved between the adhesive and the metal. Following is a brief summary of factors affecting ceramic to metal adherence and the five main theories as to why bonding occurs. (1)

A. Factors Affecting Adherence

1 - Coefficient of Expansion - The coefficients of expansion of the metal and adhesive should be as close together as possible. If the adhesive's coefficient is greater, it will tend to pop off when cooled to room temperature.

2 - Coating Composition - The composition of a ceramic controls its melting point. It would be very desirable to have the melting temperature of a ceramic adhesive the same as a temperature used in the heat treatment or aging operation of the metal.

3 - Surface Wetting - The ceramic adhesive must wet the surfaces of the metal. If wetting does not occur, the adhesive may be unable to form a reaction bond, or to obtain a mechanical grip on the metal.

4 - Reaction Time - The time at the maturing temperature is important. The time must be long enough to complete all the desired reaction. Excessive time might produce several undesirable results: (1) devitrification, (2) adhesive flowing from the joint, (3) excessive interface reaction, or others, and should be avoided.

5 - Adhesive Thickness - The cohesive strength of the ceramic adhesive may be the limiting factor of the joint. The thicker the adhesive the greater the opportunity for the production of internal faults that could lead to failure. A minimum thickness must be maintained to insure the presence of enough adhesive to complete the bonding.

6 - Effect of Adherence Oxides - Certain metallic oxides are known to improve the adherence between porcelain enamel and the base metal. Perhaps this same effect will be found in ceramic adhesives.

7 - Metal Preparation - This will involve not only metal cleaning to remove soils and oxides, but could include the deposition of a thin coating of an adherence promoting oxide or another material.

A. Factors Affecting Adherence (Continued)

8 - Evolution of Gases - Small quantities of gases or gas producers, i.e., H, N, or C are trapped in most metals upon solidification. These are either in the molecular composition or are intergranular. Upon heating, these gases tend to boil to the surface of the metal. If the ceramic adhesive forms a glassy surface before most of these gases have escaped, discontinuities in the form of micro bubbles will occur in the adhesive and/or at the interface. Gases may also originate in the medium used to carry the ceramic adhesive frit.

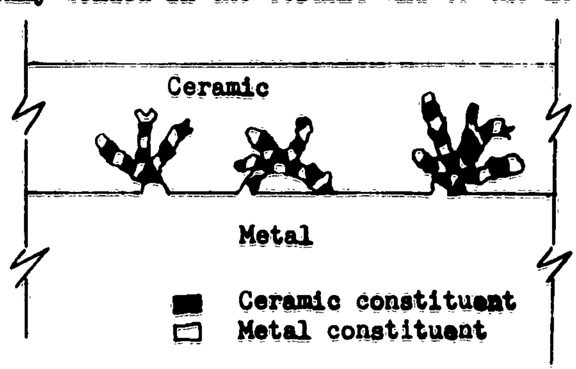
9 - Oxygen - In porcelain enameling it is generally agreed that a degree of metal oxidation occurs during the firing, and this metal oxide contributes to the adherence. The metals used in aircraft have high oxidation resistance, and it may be necessary to promote oxidation at the adhesive line in order to achieve satisfactory adherence.

10 - Firing Atmosphere - Most investigators believe the best ceramic to metal adherence is obtained in an oxidizing atmosphere. This condition would present some problems, if molybdenum were the metal being bonded.

B. Theories of Ceramic to Metal Adherence

Today, five theories are accepted for ceramic to metal adherence:

1. Metallic Dendrite Formation - During the fusion period, the metal and constituents of the ceramic unite to form interlocking crystals which are firmly bonded in the ceramic and to the metal.



2. Oxide Layer Formation - This theory states that the ceramic to metal adherence is due to the formation of thin layer of metal oxides at the interface.

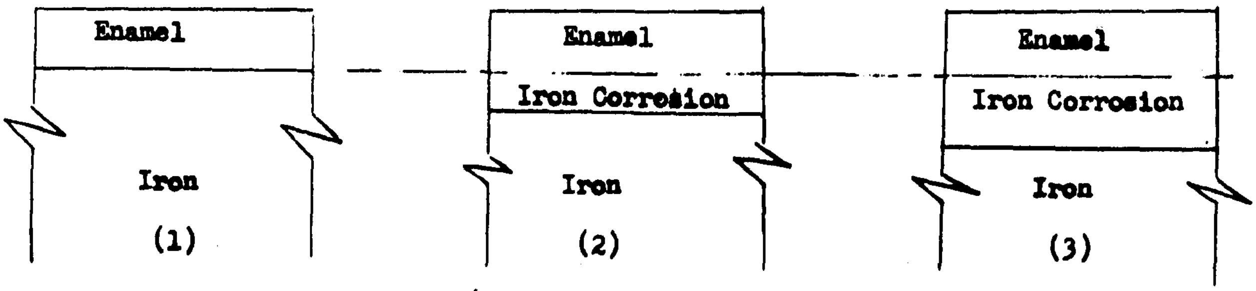
B. Theories of Ceramic to Metal Adherence (Continued)

3. Galvanic Corrosion - There is some evidence that an electrolytic reaction occurs at the ceramic-metal interface. This action seems to produce pitting and surface irregularities that permit the ceramic to "toenail" itself onto the metal. See Figure 1.

4. Mechanical Gripping - This is the same type of bonding as is found in the galvanic corrosion theory. It is here assumed the surface irregularities are formed by pickling or sandblasting prior to the application of the ceramic.

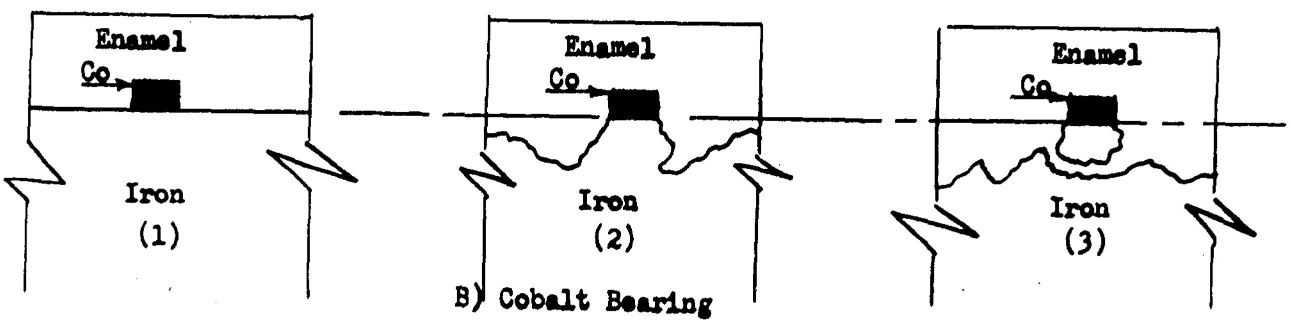
5. Chemical Bonding - Here the bonding is claimed to come from the mutual sharing of an oxygen bond between the metal and the ceramic. As the metal tries to oxidize at the higher temperatures, it successfully borrows or shares oxygen with the adherence oxides in the ceramic, thus a chemical bond is formed between the two.

DIFFERENCES IN THE CORROSIVE ATTACK ON IRON
BY COBALT FREE AND COBALT BEARING GROUND COATS



A) No Cobalt

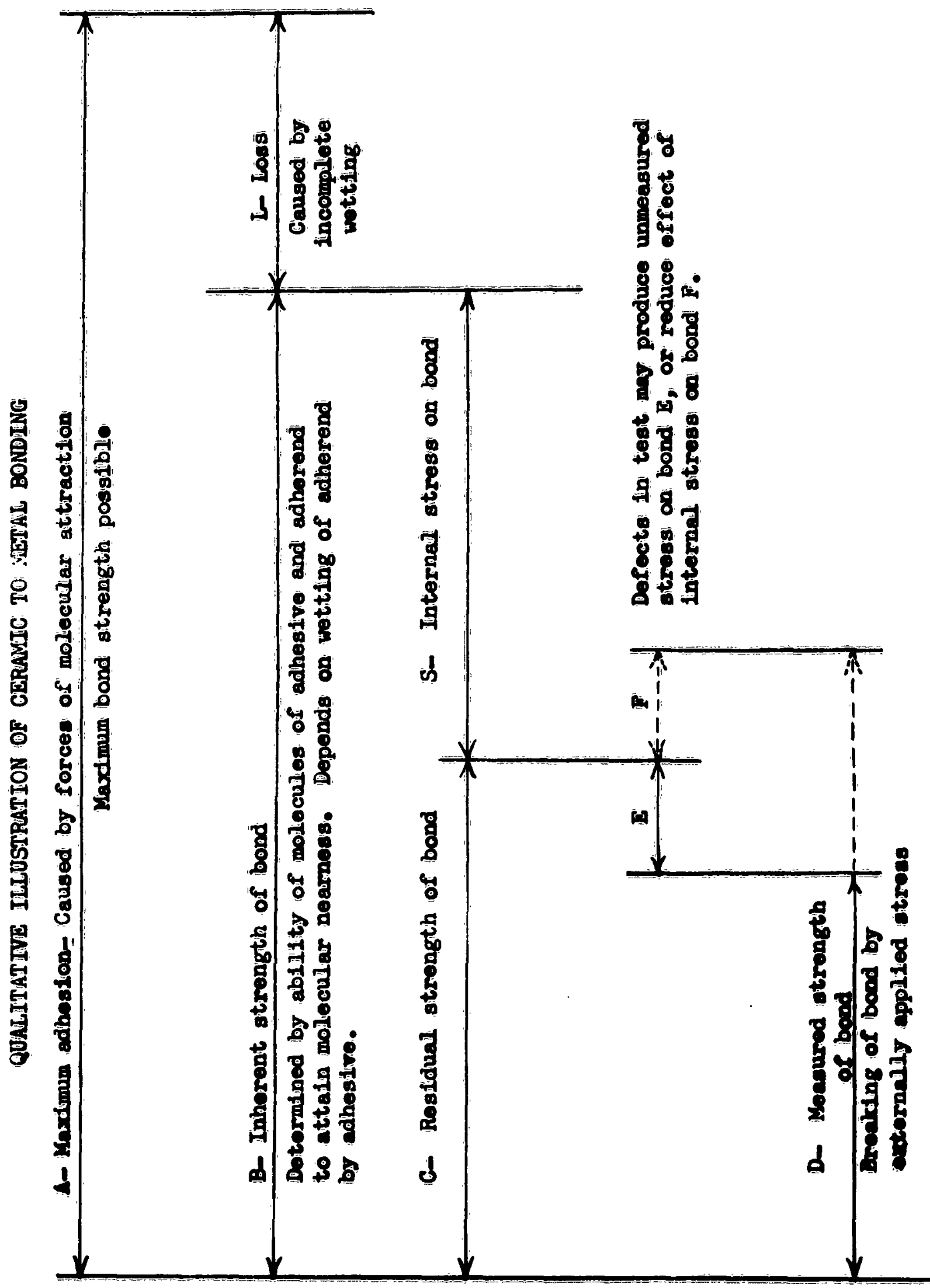
(1) 2 min. after entering furnace
(2) 4 " " " "
(3) 6 " " " "



B) Cobalt Bearing

Schematic drawing illustrating the differences in the corrosive attack on the iron by cobalt-free and cobalt bearing ground coats. The firing time increases in both sets of diagrams from left to right, the first diagram in each case indicating the interface condition shortly after the enamel fuses. From Moore, et al (4)

Figure 1



This chart is qualitative, not quantitative; no significance is attached to the relative lengths of the lines. A is always greated than B, and B greater than C. From Reinhart, F.W. (5)

Figure 2

APPENDIX II

The bibliography from "Fundamental Study of Enamel Adherence Metallic Mill Additions to Replace Adherence Oxides in Standard Enamel Ground Coats", J. E. Shoffner, Ford Motor Company Scientific Laboratory Report No. 2-48, dated January 27, 1956 (6)

This bibliography is presented to make available to the reader a listing of pertinent references on adhesion.

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